

# The Singular Effect of Disorder on Electronic Transport in Strong Coupling Electron-Phonon Systems

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We solve the disordered Holstein model in three dimensions considering the phonon variables to be classical. After mapping out the phases of the ‘clean’ strong coupling problem, we focus on the effect of disorder at strong electron-phonon (EP) coupling. The presence of even weak disorder (i) enormously enhances the resistivity ( $\rho$ ) at  $T = 0$ , simultaneously suppressing the density of states at the Fermi level, (ii) suppresses the temperature dependent increase of  $\rho$ , and (iii) leads to a regime with  $d\rho/dT < 0$ . We locate the origin of these anomalies in the disorder induced tendency towards polaron formation, and the associated suppression in effective carrier density and mobility. These results, explicitly at ‘metallic’ density, are of direct relevance to disordered EP materials like covalent semiconductors, the manganites, and to anomalous transport in the A-15 compounds.

The effect of electron-phonon (EP) interactions in weak coupling systems is well understood and can be captured within Boltzmann transport theory. However, there are materials, *e.g.* the A-15 compounds, where the electron-phonon (EP) coupling is large and the interplay of disorder and EP interaction leads to several anomalous features [1–4] in the resistivity,  $\rho$ , including a rapid rise in the residual resistivity at weak disorder, a sharp reduction in the density of states (DOS) at the Fermi level, suppression of the temperature ( $T$ ) dependence of  $\rho$  and, sometimes, a regime with  $d\rho/dT < 0$ . The conjunction of EP coupling and disorder has dramatic effects on transport in covalent semiconductors as well [5], and also controls the resistivity and optical spectra in the low  $T$  spin polarised phase of the manganites [6].

These transport anomalies, ranging across a wide variety of materials, have a common origin in the interplay of polaronic tendency (arising from strong EP coupling) with quenched disorder and thermal fluctuations. However, to capture these effects within a unified framework we need to handle the strong coupling and quenched disorder non perturbatively, and simultaneously retain the spatial correlations in the problem.

We accomplish that in this paper, solving the Holstein model in three dimensions, with arbitrary coupling and quenched disorder, through a new real space Monte Carlo (MC) technique [7] explicitly retaining the spatial correlations and localisation effects in the problem. We work with adiabatic phonons, *i.e.* treat the phonons as annealed classical variables. Apart from providing the first complete solution to this problem, our results specifically highlight: (i) the huge amplification of quenched disorder by strong EP coupling, and the dramatic increase in residual resistivity with weak disorder, (ii) the rapid suppression of the ‘thermal’ component of  $\rho(T)$  with increasing disorder, and (iii) the emergence of a regime with  $d\rho/dT < 0$ . We correlate these effects with (a) the

appearance of a weak pseudogap in the density of states and (b) the suppression of the optical spectral weight, suggest a transport phenomenology for these disordered EP systems, and compare our results with data on the disordered A-15 compounds.

*Model:* The disordered Holstein ( $d$ -H) model with spinless fermions and classical phonons is described by:

$$H = -t \sum_{\langle ij \rangle} c_i^\dagger c_j + \sum_i (\epsilon_i - \mu) n_i - \lambda \sum_i n_i x_i + H_K \quad (1)$$

The  $t$  are nearest neighbour hopping on a simple cubic lattice,  $\epsilon_i$  is the quenched binary disorder, with value  $\pm\Delta$ ,  $\mu$  is the chemical potential,  $\lambda$  is the EP interaction, coupling electron density  $n_i$  to the local distortion  $x_i$ , and the ‘restoring force’ arises from  $H_K = (K/2) \sum_i x_i^2$ . The parameters in the problem are  $\lambda/t$ ,  $\Delta/t$ , electron density  $n$ , and temperature  $T$ . We measure energy, frequency,  $T$ , *etc.* in units of  $t$ , and use  $K = 1$ .

While the strong coupling problem with one electron has been widely studied, we do not know of controlled approximations to handle strong coupling at metallic densities [8], except within dynamical mean field theory (DMFT). DMFT has been used in the adiabatic limit [9] to map out the Fermi liquid and polaronic insulator regime, and the occurrence of charge order (CO) [10]. The effect of strong EP coupling on  $\rho(T)$ , including the effect of disorder, has also been explored [11].

Our present understanding of strong coupling EP systems owe much to DMFT, but this approach has limitations in the presence of disorder. (i) Disorder and strong EP interactions reinforce each other and generate a strong local correlation between  $\epsilon_i$  and  $x_i$ , inaccessible in the statistically homogeneous description of DMFT. (ii) In such a system the conductivity can no longer be computed in terms of the single particle self energy, as in DMFT, and vertex corrections leading to localisation are crucial to recover the  $d\rho/dT < 0$  regime.

With the possibility of polaron formation, and the need to handle disorder and thermal fluctuations, MC calculation is the only unbiased method for approaching the  $d$ -H problem. However, the cost of annealing the phonon variables, via the standard exact diagonalisation based MC (ED-MC), grows rapidly with system size. For a cube with  $N = L^3$ , the ED-MC cost grows as  $N^4$ , allowing typically  $L = 4$  with current resources. We instead use a “travelling cluster approximation” (TCA) [7] for annealing the phonons. Since TCA avoids iterative diagonalisation of the electron Hamiltonian, and estimates the energy cost of MC moves based on a smaller ‘cluster’ Hamiltonian, we can access sizes upto  $12^3$  with relative ease. We use a  $4^3$  cluster for the TCA based updates. Once the phonon variables are equilibrated, we diagonalise the *full electron Hamiltonian* in the equilibrium configurations to compute [12] electronic properties. We use  $N = 8^3$ , checking with  $10^3$  for size effects. Our resistivity is in units of  $\hbar a_0 / (\pi e^2)$ , where  $a_0$  is the lattice spacing. As a rough measure,  $\rho \sim 100$  in our units is equivalent to the Mott resistivity, about  $2 - 3 \text{ m}\Omega\text{cm}$ .

Let us start with the phases. In the absence of disorder, the finite  $T$  phases can be broadly classified as (i) Fermi liquid (FL) with  $d\rho/dT > 0$ , (ii) a polaronic liquid (PL), with ‘large’ lattice distortions, no long range positional order, and  $d\rho/dT < 0$ , and (iii) the charge ordered insulator (COI) state. Unlike at  $T = 0$  [13], all the phases have finite density of states at the Fermi level. There is no ‘phase boundary’ between FL and PL and the distinction is only in terms of  $d\rho/dT$ .

For  $\lambda \lesssim 2.0$ , Fig.1.(a), the only feature in the  $n - T$  phase diagram is the occurrence of CO at  $n = 0.5$ , and the coexistence region separating the CO phase from the FL. With increasing  $\lambda$  the CO region extends down to  $n \sim 0.35$  (a chessboard pattern with ‘vacancies’ [13]), while  $T_{CO}$  at  $n = 0.5$  grows and then diminishes slowly beyond  $\lambda \simeq 3.5$ . We will discuss the CO phase in detail

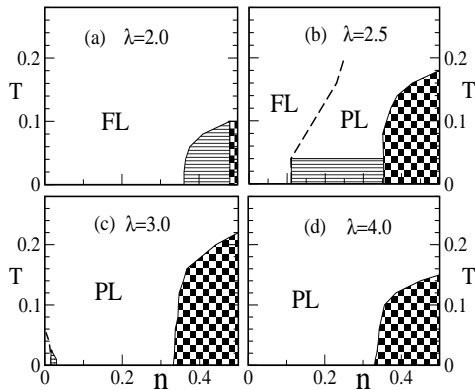


FIG. 1. Phase diagram: clean model. Panels (a)–(d). show the  $n - T$  phase diagram with increasing  $\lambda$ . The “chessboard” pattern represents a  $\{\pi, \pi, \pi\}$  CO phase, and the hatched regions indicate phase separation.

elsewhere [15]. The coexistence width at  $T = 0$  between the FL and COI increases with increasing  $\lambda$  but the temperature window for coexistence reduces since the finite  $T$  strong coupling FL itself has strong density fluctuations. By the time  $\lambda = 3.0$  the FL regime has almost vanished, the PL and COI phases dominate the phase diagram, and the system is insulating at all  $n$  and  $T$ .

The interesting regime for studying the impact of disorder and thermal fluctuations is roughly  $\lambda \sim 2 - 3$  where the system is at strong coupling but the metallic state still survives. Our studies will be focused on  $\lambda = 2.0$ , strong coupling but still metallic, and  $n = 0.3$ , and we will use  $\lambda = 1.0$  as our ‘weak coupling’ reference. To set the scale, the *single polaron* threshold is [13,14]  $\lambda_c = 3.3$ , while at  $n = 0.3$  *collective* polaronic localisation [13] sets in at  $\lambda \approx 2.5$ . We study the impact of disorder staying below this threshold.

The presence of disorder in the FL phase tends to create density inhomogeneities,  $\delta n_r$ . This effect is weak at weak disorder in an uncorrelated system but can be strongly amplified by a positive feedback if the EP coupling is large. The phenomenon is understood in the context of a single electron [5], but an understanding of the dense many electron state, and transport properties, has remained out of reach and is our main focus.

The resistivity arises from a combination of quenched disorder and lattice distortions. For a phonon configuration  $\{x\}^\alpha$ , the “potential” seen by the electrons is  $\xi_i^\alpha = \epsilon_i - \lambda x_i^\alpha$ . Subtracting out the spatial average,  $\bar{\xi}_\alpha$ , the fluctuating part is  $\delta \xi_i^\alpha \equiv \eta_i^\alpha = \epsilon_i - \lambda(x_i^\alpha - \bar{x}_\alpha)$ . Since the spatially averaged distortion does not depend on configurations, *i.e.*,  $\bar{x}_\alpha \approx \bar{x}$ , we can write  $x_i^\alpha - \bar{x}_\alpha \approx (x_i^0 - \bar{x}) + (x_i^\alpha - x_i^0)$ , where  $x_i^0$  is the  $T = 0$  distortion at  $\mathbf{R}_i$ . Let us define  $\delta x_i^0 \equiv x_i^0 - \bar{x}$  and  $\delta x_i^\alpha \equiv x_i^\alpha - x_i^0$ , so that  $\delta x_i^0$  is the  $T = 0$  distortion at a site (with respect to the spatial average), and  $\delta x_i^\alpha$  is the thermal fluctuation *about that local distortion*. In terms of these, the fluctuating background seen by the electrons is:  $\eta_i^\alpha \sim \epsilon_i - \lambda(\delta x_i^0 + \delta x_i^\alpha)$ . Averaging spatially, and over equilibrium configurations, the variance of this effective ‘disorder’ is  $\eta^2 = \langle (\epsilon_i - \lambda \delta x_i^0)^2 \rangle + \lambda^2 \langle (\delta x_i^\alpha)^2 \rangle - 2\lambda \langle (\epsilon_i - \lambda \delta x_i^0) \delta x_i^\alpha \rangle = \eta_0^2 + \eta_T^2 + \eta_{corr}^2$ , say.  $\eta_0^2$  is a measure of disorder at  $T = 0$ ,  $\eta_T^2$  is a rough measure of “thermal disorder”, and  $\eta_{corr}^2$  locally correlates the thermal fluctuations and the  $T = 0$  disorder.

In the clean FL there are no lattice distortions at  $T = 0$ , *i.e.*,  $\delta x_i^0 = 0$ , so  $\eta^2 = \lambda^2 \langle (\delta x_i^\alpha)^2 \rangle$ . The stiffness to thermal fluctuations of the  $x_i$ , at low  $T$  and weak coupling, can be roughly estimated from the energy cost  $(K/2)x_i^2 + \lambda^2 \chi(n)x_i^2$ , where  $\chi$  is the local, zero frequency, density response function of the electron system. The effective stiffness is  $K_{eff} = K + 2\lambda^2 \chi(n)$  and  $\langle (\delta x_i^\alpha)^2 \rangle \propto T/K_{eff}$ . The thermal disorder is  $\eta^2 = \eta_T^2 \sim \lambda^2 T/K_{eff}$ . Electron scattering from these fluctuations would lead to  $\rho(T) \propto \eta_T^2$ , for  $\eta_T \ll W$ , where  $W = 12t$  is the bare bandwidth, but as  $\eta_T^2$  grows the resistivity rises faster

than  $\eta_T^2$ , due to localisation corrections, unlike in DMFT. The nature of  $\eta^2$  and  $\rho(T)$  in the clean limit and  $\lambda = 1.0$ , and approximately at  $\lambda = 2.0$ , Fig.2, can be understood within the above scenario.

For  $\Delta \neq 0$  the effective disorder at  $T = 0$  arises from  $\epsilon_i$  and the induced lattice distortion:  $\eta^2 = \langle (\epsilon_i - \lambda \delta x_i^0)^2 \rangle$ . If  $\delta x_i^0$  is small, as one expects at weak disorder and weak coupling, then  $\eta^2 \approx \langle \epsilon_i^2 \rangle = \Delta^2$ , leading to  $\rho(0) \sim \Delta^2$ . We have checked this at  $\lambda = 1$  and  $n = 0.3$ , Fig.2.(a) inset, and the expected dependence clearly holds. However, when  $\lambda = 2$ , even weak disorder can create strong density inhomogeneities and induce *large* quenched distortions [5]. In this regime,  $\eta^2$  is dominated by  $\lambda^2 \langle (\delta x_i^0)^2 \rangle$ . Since  $\delta x_i^0 \sim (\lambda/K) \delta n_i^0$ , we have  $\eta^2 \approx (\lambda^4/K^2) (\delta n_i^0)^2$ . Monte Carlo annealing of the phonon variables reveals that density inhomogeneities can be strong,  $\delta n_i^0 \sim \mathcal{O}(1)$ , and the  $\lambda^4$  factor leads to a huge amplification of the external disorder. The rapid growth in  $\rho(0)$ , Fig.2.(c) inset, arises due to this. The small deviation from  $\rho(0) \propto \Delta^2$ , at small  $\Delta^2$ , is a finite size effect.

The effect of disorder and thermal fluctuations is additive in both  $\eta^2$  and  $\rho(T)$  at  $\lambda = 1.0$ , as borne out by the parallel curves in Fig.2.(a) and Fig.2.(c), *i.e.*  $\rho(T)$  obeys Mathiessens rule. However at  $\lambda = 2.0$  the same  $\Delta$  leads to *much stronger* effective disorder,  $\eta^2(T)$ , compared to  $\lambda = 1.0$ . What is more striking is that despite the  $d\eta^2/dT > 0$  in both cases the “thermal increase” in  $\rho(T)$  at  $\lambda = 2.0$  is actually *suppressed* with increasing  $\Delta$ , Fig.2.(d), leading eventually to a regime with  $d\rho/dT < 0$ , by the time  $\Delta \sim 0.5$ . We will argue that these transport anomalies at  $\lambda = 2.0$  arise from the *disorder induced polaron formation tendency*, at  $T = 0$ , and *its weakening with increasing temperature*. We next discuss

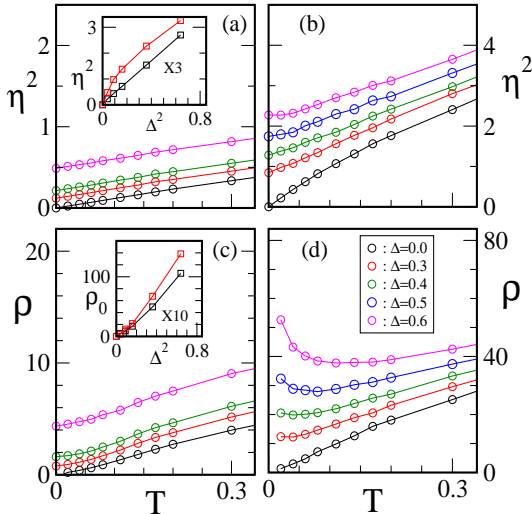


FIG. 2. (Colour online) Effective disorder,  $\eta^2$ , and resistivity,  $\rho(T)$ , at  $n = 0.3$ . (a) – (b). shows  $\eta^2$ : (a).  $\lambda = 1.0$ , (b).  $\lambda = 2.0$ . (c) – (d).  $\rho(T)$ , corresponding to the respective panel above. Inset to (a).  $\eta^2(\Delta)$  at  $T = 0$ , black  $\lambda = 1.0$ , red  $\lambda = 2.0$ , inset to (c).  $\rho(0, \Delta)$ , black  $\lambda = 1.0$ , red  $\lambda = 2.0$ .

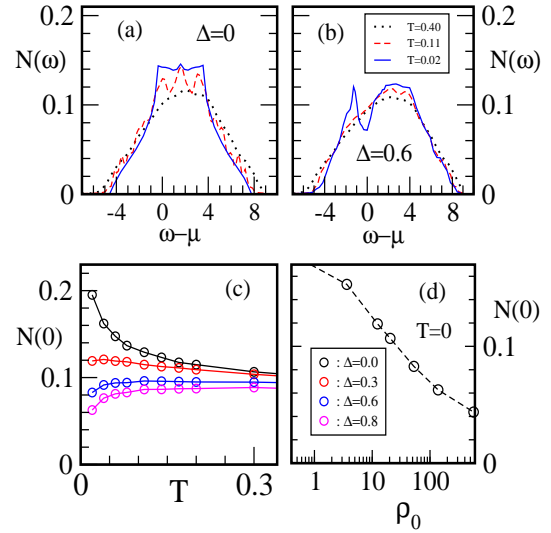


FIG. 3. (Colour online) Density of states, at  $\lambda = 2.0$ ,  $n = 0.3$ . The  $\Delta$  and  $T$  are marked on the panels. (c). Temperature dependence of  $N(0)$ , the DOS at  $\epsilon_F$ , (d). Correlation, at  $T = 0$ , between  $N(0)$  and  $\rho_0$ , varying  $\Delta$ .

the DOS and optics, and then our overall scenario.

The DOS,  $N(\omega, T)$ , is featureless in the clean problem, Fig.3.(a), even at  $\lambda = 2.0$ , typical of gradual thermal disordering of a FL. The presence of weak disorder, *e.g.*,  $\Delta = 0.6$ , Fig.3.(b), leads to the formation of a weak pseudogap around  $\omega = 0$ . The transfer of spectral weight to lower frequencies in  $N(\omega)$  suggests possible localisation of some electrons into polaronic states. The pseudogap fills quickly with increasing  $T$  and, for  $\Delta \lesssim 0.8$ , vanishes for  $T \gtrsim 0.15$ , Fig.3.(c). The suppression of DOS at  $\epsilon_F$  and the increase in residual resistivity, with increasing disorder, have a monotonic relation, Fig.3.(d).

The optical conductivity,  $\sigma(\omega)$ , in the clean problem, Fig.4.(a), is Drude like as expected of a Fermi liquid. Very weak disorder,  $\Delta = 0.3$ , Fig.4.(b), retains the Drude character, but suppresses the overall magnitude. At  $\Delta = 0.6$ , however, where anomalies were visible in  $\rho(T)$  and  $N(\omega)$ , the optical response is non Drude at all  $T$ , suggesting the existence of localised states. The “effective carrier number”  $n_{eff}(\bar{\omega}, T) = \int_0^{\bar{\omega}} \sigma(\omega, T) d\omega$ , which is a rough measure of the kinetic energy, is reduced by a factor  $\sim 10$  (at  $\bar{\omega} = 1$ ) as  $\Delta$  increases from zero to 0.6 at  $T = 0$ , Fig.4.(d). With increasing  $T$  the strongly localised particles delocalise, as borne out by the increasing  $n_{eff}(T)$ , Fig.4.(d)., and the DOS at  $\epsilon_F$ , Fig.3.(b).

We propose the following approximate “two fluid” framework to approach the results of Figs.2-4. (a). At  $T = 0$  the clean adiabatic EP problem allows only two kinds of collective states [13] at  $n = 0.3$ , (i) a band metal (with no lattice distortions) for  $\lambda \lesssim 2.5$ , or (ii) a polaronic insulator, with all electrons localised, for  $\lambda \gtrsim 2.5$ . The presence of weak disorder at  $\lambda = 2.0$  induces strong localisation of a *fraction*,  $f_{pol} = n_{pol}/n$  of charge carriers.  $n_{pol}(\Delta) \approx \int_{x_{min}}^{\infty} P(x, \Delta) dx$ , where  $P(x, \Delta)$  is the

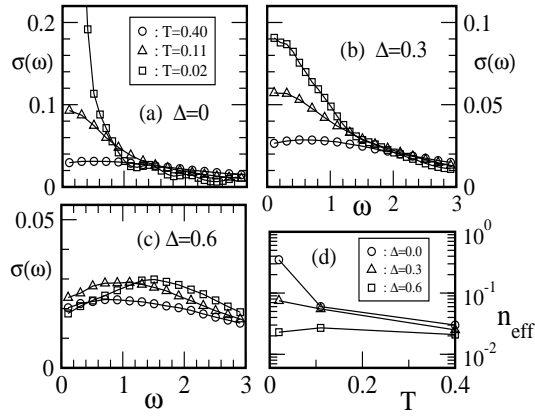


FIG. 4. Optical response: (a) – (c):  $\sigma(\omega, T)$  at  $\lambda = 2.0$  and  $n = 0.3$ . (a)  $\Delta = 0$ , (b)  $\Delta = 0.3$ , (c)  $\Delta = 0.6$ . Panel (d)  $T$  dependence of  $n_{\text{eff}}$ , the integrated low frequency optical spectral weight (see text) for various  $\Delta$ .

distribution of lattice distortions and  $x_{\min}$  is the upper edge of the clean FL peak. However, states near  $\epsilon_F$  continue to be delocalised. The fraction  $f_{\text{pol}}$  increases with increasing disorder [15], suppressing  $n_{\text{eff}}$  and  $N(0)$  and increasing  $\rho(0)$ . The fraction,  $1 - f_{\text{pol}}$ , of extended states are themselves strongly scattered by the large distortions,  $x_i$ , but still maintain ‘metallic’ conduction. (b). With increasing  $T$ , the electrons in extended states scatter off thermal fluctuations in  $x_i$  and contribute to a growing  $\rho(T)$ , while localised states close below  $\epsilon_F$  provide a “parallel” conduction channel, whose conductivity increases with increasing  $T$ . The reduction in  $d\rho/dT$  at intermediate  $\Delta$  comes from these competing tendencies. For  $\Delta \gtrsim 0.8$  there are few extended states and at low  $T$  the delocalising trend dominates. We have explicitly checked the  $T$  dependence of  $n_r$ , [15], and the density inhomogeneity, due to strongly localised electrons, weakens with increasing  $T$ . At higher  $T$ , after the polaronic effects have disappeared,  $\rho(T)$  again roughly tracks  $\eta^2$ .

Some care is needed in applying our single band adiabatic results to real materials, where (i) both the band-structure and the EP coupling could be more complicated, and (ii) the phonon frequency,  $\omega_{ph}$ , although usually  $\ll t$  is still finite. While the former will affect any quantitative comparison, the later brings in the physical effect of phonon assisted hopping, and possibly a larger conductivity from the ‘localised’ states than in the  $\omega_{ph} = 0$  limit. Such a ( $\omega_{ph} \neq 0$ ) mechanism had been studied in the limit of *strong* disorder and *weak* coupling [16] to explore the sign change in  $d\rho/dT$ . Our focus is on the complementary *strong* coupling, *weak* disorder limit.

The interplay of disorder and strong EP coupling is best documented in the ‘high  $T_c$ ’ A-15 compounds [1–3, 17]. There, in addition to ‘resistivity saturation’ [18] in the clean limit, the combination of strong EP coupling with weak disorder leads to the following effects: (i)  $\rho(0)$  increases *enormously*, in response to disorder (alpha particle damage) [1] compared to similar disorder in a ‘weak

coupling’ material, Nb, say, (ii) as  $\rho(0)$  grows, the thermal increase in  $\rho(T)$  is systematically reduced, tending to a limit  $d\rho/dT \sim 0$  at strong disorder [1], (iii) there is a degradation of  $T_c$  with increasing  $\rho(0)$ , suggesting a reduction in the DOS at  $\epsilon_F$  [2, 3]. (iv) Some other systems, *e.g.* LuRh<sub>4</sub>B<sub>4</sub> actually go over to  $d\rho/dT < 0$  with increasing  $\rho(0)$  [4]. *All the features, (i) – (iv) above, find a consistent explanation in terms of our results.* The reduction in DOS inferred from  $T_c$  degradation bears a close parallel to our correlation between  $N(0)$  and  $\rho(0)$ , Fig. 3.(d).

*Conclusion:* In this paper we solved the problem of transport in a dense, strong coupling, disordered electron-phonon system, explicitly in three dimensions. Our results reveal how disorder affects the many electron system, close to a collective polaronic instability, by strongly localising a *fraction* of electronic states, and dramatically reducing the mobility of extended states that survive near the Fermi level. We mapped out the interplay of these polaronic and extended states on transport, spectral, and optical properties, suggested a phenomenology for these class of systems, and compared our results with data on the A-15 compounds.

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